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Mass Transfer in a Closed Stirred Gas/Liquid Contactor
Part 2: The Liquid Phase Mass Transfer Coefficient k_L

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Abstract

The liquid phase mass transfer coefficient k_L for the absorption of oxygen in tap water and in ionic solutions has been calculated from the quotient of the mass transfer rate k_LS, which has been discussed in a previous article, and the specific contact area S. The specific contact area S was calculated from the formulae given by Calderbank.

It is concluded that the liquid phase mass transfer coefficient is roughly proportional to the stirrer speed. The gas fraction ϵ apparently has little influence on k_L.

INTRODUCTION

The aim of this study was to determine the liquid phase mass transfer coefficient k_L. Knowledge of the liquid phase mass transfer coefficient may be of practical interest for the design of gas/liquid reactors. This coefficient may affect the conversion rate and the selectivity of a gas/liquid reaction. In addition, knowledge of it may give information on the flow pattern around bubbles.

The coefficient is calculated by dividing the mass transfer rate k_LS by the specific contact area S. The results of the measurements of the mass transfer rate k_LS have been discussed in a previous article[1]. The specific contact area S under the same reactor conditions will be discussed in one of the sections of this article.

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THE LIQUID PHASE MASS TRANSFER COEFFICIENT k_L

The liquid phase mass transfer coefficient k_L is the rate parameter describing the mass flux from an interface to the bulk (or *vice versa*) at a given concentration difference Δc between the bulk and the interface. This coefficient, for gas absorption in a gas/liquid dispersion, is generally a function of physical properties, a characteristic dimension for the geometry, a characteristic velocity, the mobility of the interface and the coalescence rate, which in turn depends on physical properties of the interface that have not yet been defined satisfactorily. This function is represented by

$$k_L = f(D, \rho_L, \eta_L, d_b, v, \text{interface properties}) \quad (1)$$

For a bubble dispersion in a closed stirred tank, d_b and v are dependent variables. The bubble diameter is determined by the stirrer speed n, the impeller diameter D_i, the tank diameter T, physical properties such as the liquid and gas viscosities η_L and η_G and the liquid and gas densities ρ_L and ρ_G, the gas fraction ϵ and the coalescence rate.

The characteristic velocity is determined by the terminal velocity and a characteristic velocity for the intensity of turbulence around a bubble. The terminal velocity is a function of the bubble diameter d_b, the acceleration due to gravity g, the density difference $\rho_L - \rho_G$, the liquid viscosity η_L and physical properties which determine the mobility of the interface. The characteristic turbulence velocity is a function of the stirrer speed n, the impeller diameter D_i, the tank diameter T, the liquid density ρ_L and the position in the tank.

Thus, by elimination of d_b and v in eqn. (1) we obtain

$$k_L = f(D, \rho_L, \rho_G, \eta_L, \eta_G, g, n, D_i, T, \epsilon, \text{the physical properties determining the mobility of the interface and the coalescence rate, the position in the tank}) \quad (2)$$

One might expect *a priori* that the mass transfer coefficient will depend on the rate of coalescence for two reasons:

(1) coalescence may give rise to larger bubbles that have a more mobile interface (larger k_L); and
(2) rapid coalescence causes rapid re-dispersion which means rapid surface renewal (larger k_L).

Calculation of k_L

The previous article[1] reports an investigation in which the mass transfer rate $k_L S$ was determined in a closed stirred tank as a function of the stirrer speed n, the tank diameter T and the ratio of the stirrer diameter to the tank diameter D_i/T, in two solutions:

(a) tap water, in which the coalescence rate is very high, and
(b) ionic solutions, in which the coalescence rate is very low.

The above-mentioned mass transfer rate is a mean value for the whole tank. It may be expected that the mass transfer rate is a function of the position in the tank, since the specific contact area S as well as the liquid phase mass transfer coefficient k_L may be functions of the position in the tank.

In view of these complications we were only able to calculate an average liquid phase mass transfer coefficient $k_{L,m}$, defined as the ratio of the over-all (or mean) value $\overline{k_L S}$ of $k_L S$ and the over-all value $\bar{S}$ of the specific contact area:

$$k_{L,m} = \overline{k_L S}/\bar{S} \quad (3)$$

The determination of the specific contact area will be discussed next.

THE SPECIFIC CONTACT AREA S

Definition of S

The specific contact area S in a gas/liquid dispersion is defined as the interfacial contact area of all the bubbles in a volume element ΔV, divided by the volume of that element. We may distinguish between the over-all specific contact area $\bar{S}$ for the whole reactor with volume V and the local specific contact area $S(i)$ for a small volume element $\Delta V(i)$. In practice $\Delta V(i)$ cannot be infinitely small—it must be larger than the mean bubble diameter. The main difficulty in the determination of the over-all specific contact area $\bar{S}$ in a stirred tank reactor is that in such a reactor the specific contact area depends strongly, in general, on the position in the tank, because the specific contact area S is a function of the gas fraction ϵ and the Sauter mean diameter $d_{3,2}$:

$$S = 6\epsilon/d_{3,2} \quad (4)$$

The dependence on the position in the tank may be a consequence of a variation in the local gas fraction and/or the local Sauter mean diameter.

Determination of S

The local interfacial contact area $S(i)$ can be determined directly (with certain restrictions) by the following techniques:

(1) a light transmission technique, and
(2) a light reflection technique.

It can be determined indirectly by a determination of the local gas fraction $\epsilon(i)$ and the local Sauter mean diameter $d_{3,2}(i)$.

The over-all interfacial contact area over the whole tank can be determined with the chemical technique (absorption accompanied by a fast chemical reaction) but this technique cannot be used for fast-coalescing systems, since the presence of chemical compounds usually reduces the coalescence rate. Apart from this, in fast-coalescing systems the value of the specific contact area depends strongly on the position in the tank, which reduces the meaning of an average value.

Correlations for open and closed stirred tanks

The interfacial area in a stirred tank reactor has been determined by Calderbank[2] in a 100 l and a 5 l tank by using a light transmission technique. Calderbank distinguished two classes of solutions:

(a) pure solutions, in which the rate of coalescence is very high, and
(b) ionic solutions, in which the rate of coalescence is very low.

He used an open tank reactor for the determination of the interfacial area in pure solutions and verified his results with a few measurements in a closed tank.

In the 100 l open tank about 100 measurements were made of the local interfacial area $S(i)$ and the local hold-up $\epsilon(i)$, whereas in the 5 l tank only one measurement was made. From the local values of the interfacial area he calculated a mean value $\bar{S}$ for the whole tank and from the local values of $\epsilon(i)$ he calculated a mean value $\bar{\epsilon}$ for the whole tank. Then the Sauter mean diameter was calculated with the relation (4).

Calderbank proposed the following empirical relation for the mean value of the Sauter diameter of bubbles in pure solutions:

$$d_{3.2} = 4.15 \frac{\sigma^{0.6}}{(P/V)^{0.4}\rho_L{}^{0.2}} \epsilon^{0.5} + 0.0009 \qquad (5)$$

He used only a closed tank for the determination of the over-all specific contact area $\bar{S}$ in ionic solutions, and proposed the following empirical relation for the Sauter mean diameter:

$$d_{3.2} = 2.25 \frac{\sigma^{0.6}}{(P/V)^{0.4}\rho_L{}^{0.2}} \epsilon^{0.4} \left(\frac{\eta_G}{\eta_L}\right)^{0.25} \qquad (6)$$

We have used these two formulae to recalculate the over-all specific interfacial contact area $\bar{S}$ for the same conditions at which values of the mass transfer rate $k_L S$ were determined.

DISCUSSION OF RESULTS

For a number of experimental conditions, the mean value of the liquid phase mass transfer coefficient $k_{L,m}$ has been calculated according to eqn. (3) from our experimental values of $\overline{k_L S}$ and from the predicted values of $\bar{S}$. The following variables were taken into account.

For a tank diameter T of 0.6 m and ratios D_i/T of 0.3, 0.35 and 0.4, the stirrer speed was varied between 3 and 13 s^{-1}.

For a tank diameter T of 0.19 m and ratios D_i/T of 0.3, 0.35 and 0.4, the stirrer speed was varied between 11.6 and 33.3 s^{-1}.

Three different aqueous solutions were used:

(a) tap water,
(b) a solution of 0.6 kmol NaCl/m^3 and
(c) a solution of 0.8 kmol Na_2SO_3 or Na_2SO_4/m^3.

Solution (c) was only used in the smaller vessel.

Tap water

For tap water it was found that $k_{L,m}$ increased roughly in proportion to the stirrer speed n. The influence of D_i and T could be taken into account by correlating $k_{L,m}$ with the group $nD_i^2/T^{1.6}$. In Fig. 1 a plot has been made of $k_{L,m}$ as a function of

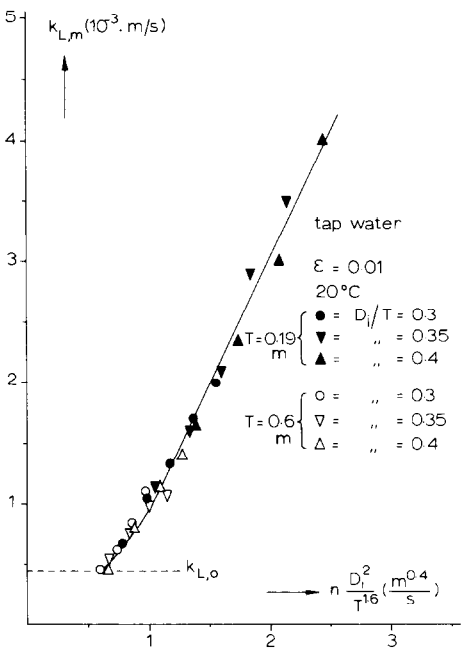

Fig. 1. The mean value of the liquid phase mass transfer coefficient ($k_{L,m}$) in tap water as a function of the group $n D_i^2/T^{1.6}$.

this group. The dotted line in this graph represents the value of the liquid phase mass transfer coefficient $k_{L,0}$, which has been determined by Calderbank and Moo Young[3] for free rising bubbles with a mobile interface in a swarm and in a column without stirring. It may be expected that the liquid phase mass transfer coefficient for an analogous gas/liquid dispersion in a stirred tank will not decrease below $k_{L,0}$. The minimum value of $k_{L,m}$ which we have measured is indeed equal to $k_{L,0}$.

The surprising increase in $k_{L,m}$ may be a consequenc of

(1) an increased coalescence and re-dispersion rate, and

(2) an increased surface renewal rate as a consequence of the increase in the intensity of turbulence.

A decrease might have been expected as a consequence of a decrease in the mean bubble diameter.

In view of the existence of these various effects, it will be impossible to estimate their relative influence on $k_{L,m}$ and it is not possible to give even a qualitative explanation of these results.

Aqueous solution of 0.6 kmol NaCl/m³

The empirical correlation for the mass transfer rate in an aqueous solution of 0.6 kmol NaCl/m³ is given in the previous article[1]:

$$\overline{k_L S} = 0.11 \left(\frac{n^3 D_i^5}{T^3}\right)^{0.7} \left(\frac{D_i}{T}\right)^{0.7} \left(\frac{1}{T}\right)^{0.35} \qquad (7)$$

An expression for the specific contact area $\bar{S}$ can be derived from eqns. (5) and (6):

$$\bar{S} = c_1 \left(\frac{n^3 D_i^5}{T^3}\right)^{0.4} \qquad (8)$$

where

$$c_1 = 6 \left(\frac{4 \times 5.8}{\pi}\right)^{0.4} \frac{\epsilon^{0.6}}{2.25} \left(\frac{\eta_L}{\eta_G}\right)^{0.25} \left(\frac{\rho_L}{\sigma}\right)^{0.6} \qquad (9)$$

The following relation is obtained from the quotient of the mass transfer rate $k_L S$ and the specific contact area $\bar{S}$:

$$k_{L,m} = \frac{0.11}{c_1} \left(\frac{n^3 D_i^5}{T^3}\right)^{0.3} \left(\frac{D_i}{T}\right)^{0.7} \left(\frac{1}{T}\right)^{0.35} \qquad (10)$$

Thus $k_{L,m}$ is not a unique function of the power dissipation per unit mass; it is also a function of the ratio D_i/T and the tank diameter T.

Levins and Glastonbury[4] also reported that the liquid phase mass transfer coefficient $k_{L,m}$ for the dissolution of solid particles is a function of the power dissipation per unit mass and the ratio D_i/T.

Na_2SO_3 solution

In Fig. 2 the effect of the stirrer speed on the liquid-phase mass transfer coefficient of a bubble dispersion in an aqueous solution of 0.8 kmol Na_2SO_3/m^3 is compared with the results of other authors. Reith[5] concluded that $k_{L,m}$ was independent of the stirrer speed (line 1) and van Dierendonck[6] concluded that $k_{L,m}$ decreases with increasing stirrer speed (line 2), whereas our results show an increase of k_L with stirrer speed (line 3).

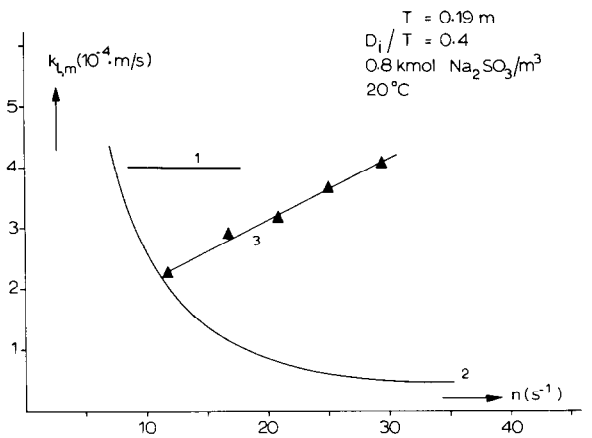

Fig. 2. The mean value of the liquid phase mass transfer coefficient ($k_{L,m}$) in a Na_2SO_3-solution as a function of the stirrer speed and in comparison with the results of other authors; 1: Reith[5]; 2: van Dierendonck[6]; 3: our results.

However, it should be noted that our results are not really comparable with those of Reith and van Dierendonck, since the effect of the gas hold-up is not shown. In the experiments of these two authors the gas hold-up, which is a dependent variable, increased from about 10% by volume to about 40% by volume if the stirrer speed increased from 15 to 33 s^{-1}. In our experiments the gas hold-up was an independent variable and it was kept constant at 1% by volume.

To demonstrate the effect of the gas hold-up we determined k_L as a function of the gas hold-up at one stirrer speed. These values of k_L were determined by

(1) measuring $k_L S$ by means of the sulphite method—since large gas fractions can only be completely dispersed at high stirrer speeds, the measurements were performed only at one stirrer speed of 29.2 s^{-1}—and

(2) calculating the corresponding values of S using eqns. (4) and (6). The specific power consumption as a function of the gas fraction was determined in a separate experiment[7].

The results are given in Table 1. They show that the liquid phase mass transfer coefficient apparently does not depend much on the gas fraction ϵ. It should be noted that $d_{3,2}$ varies together with ϵ. It is not known which of these two variables has the greatest effect on $k_{L,m}$.

TABLE 1

ϵ	$n(s^{-1})$	$k_L S(s^{-1})$	$k_{L,m}$ (m/s)	$d_{3.2}$ (mm)	$\bar{S}(m^{-1})$
0.01	29.2	0.35	4.2×10^{-4}	0.07	835
0.025	29.2	0.65	4.6×10^{-4}	0.11	1410
0.05	29.2	0.78	3.8×10^{-4}	0.15	2050
0.1	29.2	1.07	3.9×10^{-4}	0.22	2740
0.2	29.2	1.3	3.8×10^{-4}	0.35	3420

CONCLUSIONS

(1) The liquid phase mass transfer coefficient for absorption of oxygen in tap water and in ionic solutions increases (above the minimum stirrer speed needed for complete suspension of the gas phase) with increasing stirrer speed.

(2) The results for tap water could be well correlated if they were plotted against the group $nD_i^2/T^{1.6}$.

The results in an aqueous solution of 0.6 kmol NaCl/m^3 for k_L are

$$k_{L,m} = 3.25 \times 10^{-4} \left(\frac{n^3 D_i^5}{T^3}\right)^{0.3} \left(\frac{D_i}{T}\right)^{0.7} \left(\frac{1}{T}\right)^{0.35}$$

if $\epsilon = 0.01$; the constant has the units $m^{0.75}/s^{1.9}$.

(3) Larger impeller diameters D_i apparently result in larger values for the liquid phase mass transfer coefficient k_L at the same power dissipation per unit volume.

(4) In the range 0.01–0.20, the gas fraction ϵ apparently has little influence on $k_{L,m}$.

NOMENCLATURE

$d(i)$ diameter of the ith class of size distribution, m
d_b diameter of bubble, m
$d_{3.2}$ Sauter diameter, m
D diffusion coefficient, m^2/s
D_i impeller diameter, m
g acceleration of gravity, m/s^2
i integer
k_L liquid phase mass transfer coefficient, m/s
$k_{L,0}$ liquid phase mass transfer coefficient for free rising bubbles with a mobile interface, m/s
$k_{L,m}$ liquid phase mass transfer coefficient (mean value), m/s
n impeller speed, 1/s
P dissipated power in the tank, Nm/s
S specific contact area, 1/m
$S(i)$ specific contact area of volume element, $\Delta V(i)$, 1/m
T tank diameter, m
v velocity, m/s
V volume of the tank, m^3
$\Delta V(i)$ volume element of the tank ($d_b \ll \Delta V(i) \ll V$), m^3

Greek symbols

ϵ the gas fraction in the tank
η_G dynamic viscosity of the dispersed phase Ns/m^2
η_L dynamic viscosity of the continuous phase, Ns/m^2
ρ_G density of the dispersed phase, kg/m^3
ρ_L density of the continuous phase, kg/m^3
σ surface tension, N/m

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